THE TIME DEVELOPMENT AND TEMPERATURE EFFECTS ON HIGHLY CORRELATED MOTIONS OF ATOMS IN SUPERCOOLED FLUIDS

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We have carried out molecular dynamics simulations on model supercooled fluids, which are composed of equal numbers of two different species interacting through soft-core potentials, 10,000 atoms in two dimension and 500 atoms in three dimension. Keeping the reduced number density constant i.e., $\rho^* = 0.8$, the coupling constant Γ_{eff} and the reduced temperature T^* are varied as below.

Γ_{eff}	0.8	1.0	1.2	1.4
T^*	9.685	2.539	0.8509	0.3372

Analyzing the mean square displacements and non-Gaussian parameters, it has been realized that two low-Γ_{eff} states, $\Gamma_{eff} = 0.8$ and 1.0, correspond to liquid states, while two high-Γ_{eff} states, $\Gamma_{eff} = 1.2$ and 1.4 correspond to a supercooled liquid and a glass state, respectively.

Fig.1

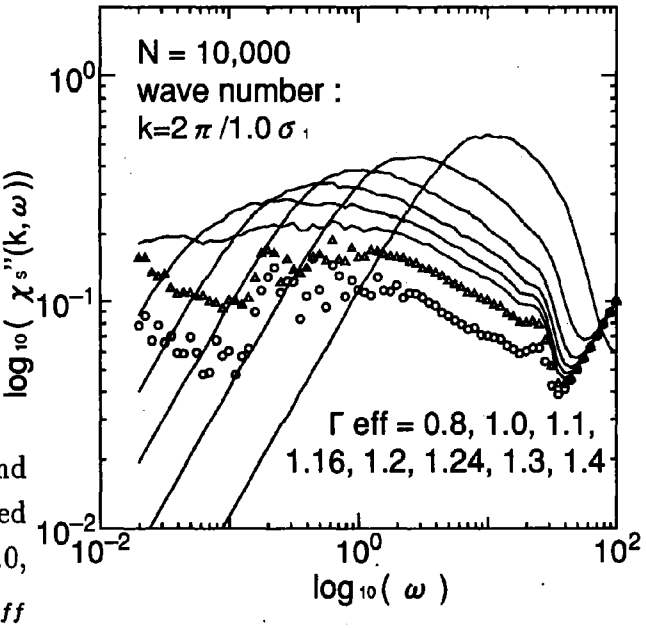

Fig.2

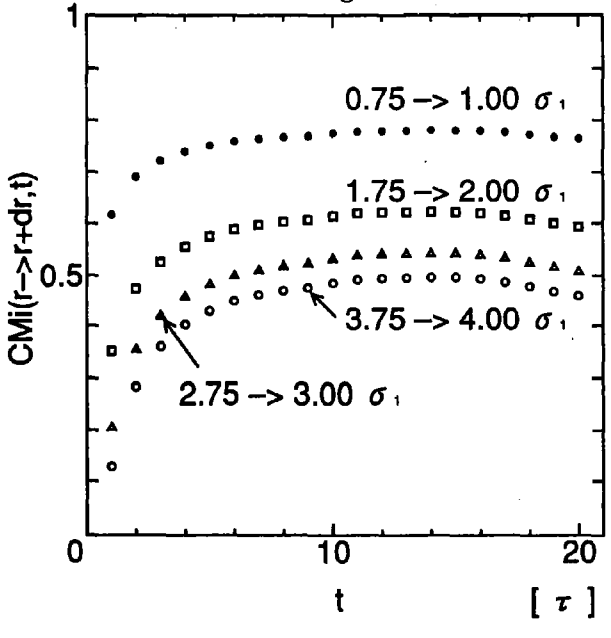

Figure 1 shows the imaginary part of the generalized susceptibility (self-part) $\chi_s''(\mathbf{k},\omega)$[1] at various Γ_{eff}'s for species 1. The reduced wave number is fixed at $k = 2\pi/1.0$(in units of σ_1^{-1}). Curves with the highest to the lowest peak corresponds to $\Gamma_{eff} = 0.8$ to 1.4.

We introduce the correlated motion coefficient[2]

$$CM_i(r_c,t) = \sum_{r_{ij}\le r_c}^{N_i} \Delta\mathbf{r}_i(t)\cdot\Delta\mathbf{r}_j(t)/\overline{\Delta r^2(t)}N_i,$$

where $\Delta\mathbf{r}_i(t)$ is the displacement vector of the i-th atom for an elapsed time t, $\overline{\Delta r^2(t)}$ is the mean square displacement for an elapsed time t, and N_i is the number of atoms within the distance r_c centered at the i-th atom for $t = 0$.

In Fig. 2, $CM_i(r \to r+dr, t)$ averaged over i and representing $CM_i(r+dr,t)-CM_i(r,t)$ is shown for $\Gamma_{eff} = 1.4$. Note that this is exactly zero for ideal gases. So far as the time development of CM_i is concerned, $CM_i(r \to r+dr, t)$ rapidly increases before 5 τ. The remarkable phenomenon is seen in that high values of CM_i range over fairly long distances[2].

Fig.3

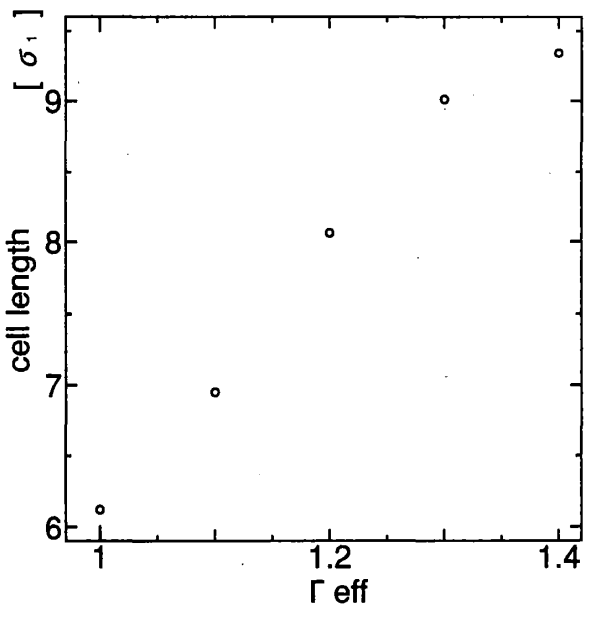

Figure 3 shows the temperature effect of CM_i[3]. The area of domains of highly correlated motions spread wider in glass states than in liquid states. The correlation length looks almost saturated near the glass transition temperature, in which the curve of figure 1 ($\chi_s''(\mathbf{k},\omega)$) has a peak which can be identified as the β peak. All these results indicate the relation between the correlated motion and the β peak.

The system size dependence was also studied in our model supercooled fluids via molecular dynamics (MD) simulation. For such purposes we have also calculated a system of 100 atoms in two dimension and a system of 500 atoms in three dimension.

One possibile view about the glass transition is that it occurs at when the area of highly correlated motion become saturated.

Fig.4

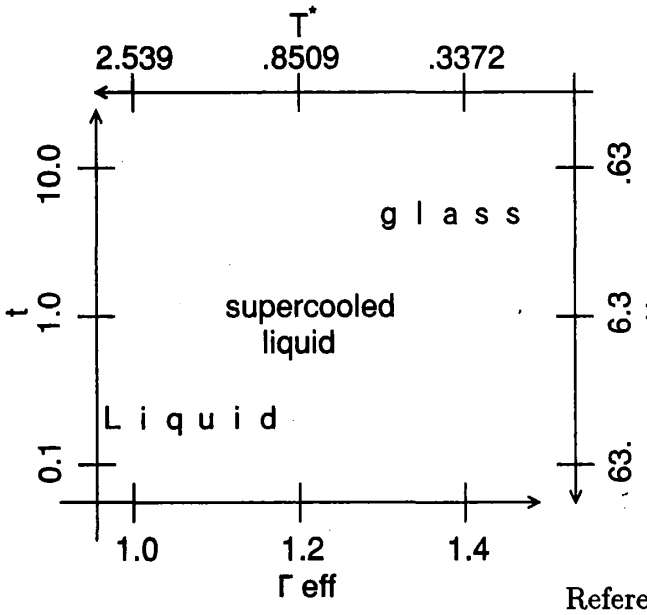

Figure 4 shows the range of time and temperature in which we are inerested and in which we have analyzed dynamical properties of atoms; time scales of 0.1 τ and $\Gamma_{eff} \leq 1.0$ corresponding to liquid states, and time scales of 5.0 τ and $\Gamma_{eff} \geq 1.3$ corresponding to glass states.

We will also discuss in detail about our MD simulation and results obtained.

References

[1] J.Matsui, H.Miyagawa, T.Muranaka, K.Uehara, T.Odagaki and Y.Hiwatari, Molecular Simulation **12**, 305 (1994).

[2] T.Muranaka and Y.Hiwatari, Phys. Rev. E **51**, R2735 (1995).

[3] T.Muranaka and Y.Hiwatari, submitted to Molec.Simu.